



RECEIVED  
NOV 05 2003  
GROUP 1700

[Document] Specification

[Title of the Invention] Process For Preparing Detergen  
Particles

[Claims]

[Claim 1] A process for preparing detergent particles, comprising the steps of

(I): mixing a base particle [(a) component] having an average particle size of from 150 to 500  $\mu\text{m}$ , a bulk density of 400 g/L or more and supporting ability of 20 mL/100 g or more, with a surfactant composition comprising a nonionic surfactant and an immobilization agent of the surfactant, wherein the surfactant composition [(b) component] comprises the immobilization agent in an amount of 1 to 100 parts by weight, based on 100 parts by weight of the nonionic surfactant, under conditions such that the mixing is carried out at a mixing ratio of the 15 to 100 parts by weight of said (b) component, based on 100 parts by weight of said (a) component, and the maximum temperature of the mixture of said (a) component with said (b) component during the period of from the initiation of mixing to the termination of mixing becomes a temperature of the pour point of said (b) component or higher; and

(II): mixing the mixture obtainable in step (I) with fine powder [(c) component],

wherein the detergent particles have a degree of particle growth of 1.5 or less as calculated by the following equation:

$$\text{Degree of Particle Growth} = \frac{\text{Average Particle Size of Final Detergent Particles After Termination of Step (II)}}{\text{Average Particle Size of (a) Component}} \text{ and}$$

a bulk density of 500 g/L or more.

[Claim 2] The process according to claim 1, wherein said (b) component further comprises 20 to 200 parts by weight of an anionic surfactant having sulfate group or sulfo group, based on 100 parts by weight of the nonionic surfactant.

[Claim 3] The process according to claim 1 or 2, wherein in step (I), mixing is initiated after raising the temperature of each of (a) component and (b) component to a temperature of a pour point of said (b) component or higher.

[Claim 4] The process according to any one of claims 1 to 3, wherein in step (I), mixing is carried out with maintaining a temperature of a mixture of said (a) component and said (b) component at a pour point of said (b) component or higher during a period between an initiation of mixing and a termination of mixing.

[Claim 5] The process according to any one of

claims 1 to 4, wherein the detergent particles have a dissolution rate of 90% or more, under conditions where the resulting detergent particles are supplied in water at 5°C; stirred for 60 seconds under the stirring conditions that 1 g of the detergent particles are supplied to a one-liter beaker having an inner diameter of 105 mm which is charged with one-liter of hard water having 71.2 mg  $\text{CaCO}_3/\text{L}$ , wherein a molar ratio of Ca/Mg is 7/3, and stirred with a stirring bar of 35 mm in length and 8 mm in diameter at a rotational speed of 800 rpm; and filtered with a standard sieve having a sieve-opening of 74  $\mu\text{m}$  as defined by JIS Z 8801, wherein the dissolution rate of the detergent particles is calculated by the equation:

$$\text{Dissolution Rate (\%)} = \{1 - (T/S)\} \times 100$$

wherein S is a weight (g) of the detergent particles supplied; and T is a dry weight (g) of insoluble remnants of the detergent particles remaining on the sieve when an aqueous solution prepared under the above stirring conditions is filtered with the sieve, wherein drying conditions for the insoluble remnants are keeping at a temperature of 105°C for 1 hour, and then in a desiccator with a silica gel at 25°C for 30 minutes.

[Detailed Description of the Invention]

[0001]

[Technical Field to Which the Invention Pertains]

The present invention relates to a process for preparing uni-core detergent particles supporting a surfactant composition on a base particle.

[0002]

[Prior Art]

As a technique for formulating a nonionic surfactant to a non-liquid detergent such as a powder detergent, Japanese Patent Laid-Open No. Hei 10-176200 discloses a process for preparing nonionic detergent granules comprising previously mixing a nonionic surfactant, a water-soluble, nonionic, organic compound having a melting point of 45°C or more, and an acid precursor of a fatty acid or the like, to give a liquid mixture; and granulating a mixture of the resulting liquid mixture with raw materials for detergents, with tumbling with an agitation mixer, thereby increasing its bulk density. However, the fatty acid reacts in the nonionic surfactant by contacting the liquid mixture with an alkalizing agent, thereby causing gelation (nonionic surfactant/soap gel). Therefore, the surfactant composition comprising a nonionic surfactant is less likely be absorbed and embedded in powder raw materials having supporting ability, and the powder raw materials are aggregated with the gelated product acting as a binder, whereby the granulation proceeds. In other words, in the above

process, the granulation proceeds without sufficiently exhibiting its supporting ability, even when using powder raw materials having supporting ability in the powder raw materials, so that a large amount of the surfactant cannot be formulated. When a large amount of the surfactant is to be formulated, granules having sizes outside of the desired particle size ranges are formed, so that it tends to be disadvantageous in the dissolubility.

[0003]

[Problems to Be Solved by the Invention]

Accordingly, a problem of the present invention is to provide in a process for preparing detergent particles comprising a nonionic surfactant, the process for preparing detergent particles having a large surfactant content in a simple preparation process, excellent dissolubility, and being excellent in the suppression of exudation of the nonionic surfactant and in the anti-caking property.

[0004]

[Means to Solve the Problems]

Specifically, the present invention relates to a process for preparing detergent particles, comprising the steps of

(I): mixing a base particle [(a) component] having an average particle size of from 150 to 500  $\mu\text{m}$ , a bulk

density of 400 g/L or more and supporting ability of 20 mL/100 g or more, with a surfactant composition comprising a nonionic surfactant and an immobilization agent of the surfactant, wherein the surfactant composition [(b) component] comprises the immobilization agent in an amount of 1 to 100 parts by weight, based on 100 parts by weight of the nonionic surfactant, under conditions such that the mixing is carried out at a mixing ratio of the 15 to 100 parts by weight of the (b) component, based on 100 parts by weight of the (a) component, and the maximum temperature of the mixture of the (a) component with the (b) component during the period of from the initiation of mixing to the termination of mixing becomes a temperature of the pour point of the (b) component or higher; and

(II): mixing the mixture obtainable in step (I) with fine powder [(c) component], wherein the detergent particles have a degree of particle growth of 1.5 or less as calculated by the following equation:

$$\text{Degree of Particle Growth} = \frac{\text{Average Particle Size of Final Detergent Particles After Termination of Step (II)}}{\text{Average Particle Size of (a) Component}} \text{ and}$$

a bulk density of 500 g/L or more.

[0005]

[Modes for Carrying out the Invention]

1. (a) Component

(a) Component refers to a particle (base particle), the particle having an average particle size of from 150 to 500  $\mu\text{m}$ , a bulk density of 400 g/L or more, and a supporting ability of 20 mL/100 g or more.

[0006]

(a) Component has an average particle size of from 150 to 500  $\mu\text{m}$ , preferably from 180 to 350  $\mu\text{m}$ , from the viewpoint of obtaining detergent particles excellent in the dissolubility and the flowability. The bulk density is 400 g/L or more, preferably 500 g/L or more, from the viewpoint of the compression of the detergent particles. Also, the bulk density is preferably 1500 g/L or less, more preferably 1200 g/L or less, from the viewpoint of the dissolubility. The supporting ability is 20 mL/100 g or more, preferably 30 mL/100 g or more, more preferably 40 mL/100 g or more, from the viewpoint of enhancing the support of a surfactant composition. The "supporting ability" refers to the ability of the base particle to support a liquid component such as a surfactant inside and on the surface of the particle. When the supporting ability is within this range, the aggregation of (a) components is suppressed, thereby making it favorable for

maintaining the uni-core property owned by the detergent particle in the detergent particles.

[0007]

In addition, the (a) component is preferably a harder substance, from the viewpoint of suppressing the breakdown of the (a) component during mixing in the steps (I) and (II). Concretely, when expressed by the particle strength, that for (a) component is preferably 100 kg/cm<sup>2</sup> or more, more preferably 200 kg/cm<sup>2</sup> or more, especially preferably 300 kg/cm<sup>2</sup> or more. (a) Component showing the particle strength as described above can be obtained by, for example, spray-drying an aqueous slurry comprising a detergent builder and the like.

[0008]

The average particle size of (a) component is measured by vibrating a sample with each of standard sieves according to JIS Z 8801 for 5 minutes, and thereafter determining from a weight percentage depending upon the size openings of the sieves. The bulk density of (a) component is measured by a method according to JIS K 3362.

The supporting ability of (a) component is measured by the following method.

A cylindrical mixing vessel of an inner diameter of 5 cm and a height of 15 cm which is equipped with



agitation impellers in the inner portion thereof is charged with 100 g of a sample. With stirring the agitation impellers at 350 rpm, linseed oil at 25°C is supplied into the vessel at a rate of 10 mL/min. The supporting ability is defined as an amount of linseed oil supplied when the agitation torque reaches the highest level.

[0009]

The particle strength is measured by the following method.

A cylindrical vessel of an inner diameter of 3 cm and a height of 8 cm is charged with 20 g of a sample, and the sample-containing vessel (manufactured by Tsutsui Rikagaku Kikai K.K., "Model TVP1" tapping-type close-packed bulk density measurement device; tapping conditions: frequency of 36 times/minute, free fall from a height of 60 mm) is tapped for 30 times. A sample height immediately after the tapping operation is measured, and defined as an initial sample height. Thereafter, an entire upper end surface of the sample kept in the vessel is pressed at a rate of 10 mm/min with a pressing machine to take measurements for a load-displacement curve. The slope of the linear portion having a displacement ratio of 5% or less in the curve is multiplied by an initial sample height, and the resulting product is divided by a pressed

area, to give a quotient which is defined as particle strength.

[0010]

(a) Component can be obtained by, for example, drying an aqueous slurry comprising a detergent builder and the like. Among them, the particle obtainable by spray-drying the aqueous slurry is preferable from the viewpoint of having desired property values. In addition, when (a) component is a spray-dried particle, it is more preferable because the detergent particles obtainable by the process of the present invention can actually have a fast dissolubility. The "fast dissolubility" refers to a property that the detergent particles have a dissolution rate described below of 90% or more.

[0011]

(a) Component can be prepared, for example, by spray-drying an aqueous slurry comprising a water-insoluble inorganic compound, a water-soluble polymer and a water-soluble salt, in which the contents of each of the components are respectively from 20 to 90% by weight, from 2 to 30% by weight, and from 5 to 78% by weight, on the basis of solid ingredients in the aqueous slurry. Within the above compositional ranges, the average particle size, the bulk density, the supporting ability and the particle strength can be controlled by adjusting the drying process

and the drying conditions. The contents of the water-insoluble inorganic compound, the water-soluble polymer and the water-soluble salt in the aqueous slurry are respectively more preferably within the ranges of from 30 to 75% by weight, from 3 to 20% by weight, and from 10 to 67% by weight, especially preferably within the ranges of from 40 to 70% by weight, from 5 to 20% by weight, and from 20 to 55% by weight, on the basis of solid ingredients in the aqueous slurry.

[0012]

The water-insoluble inorganic compound includes crystalline or amorphous aluminosilicates; silicon dioxide, hydrated silicate compounds, clay compounds such as perlite and bentonite, and the like. The water-soluble polymer includes carboxylic acid-based polymers, carboxymethyl cellulose, water-soluble starches, sugars, and the like. The water-soluble salts include water-soluble inorganic salts representatively exemplified by alkali metal salts, ammonium salts or amine salts, each having carbonate radical, hydrogencarbonate radical, sulfate radical, sulfite radical, hydrogensulfate radical, chloride radical, phosphate radical, or the like; and water-soluble organic salts having low molecular weights such as citrates and fumarates, and the like.

[0013]

## 2. (b) Component

(b) Component is a surfactant composition comprising a nonionic surfactant and an immobilization agent of nonionic surfactant. The nonionic surfactant in (b) component is preferably those having a melting point of 30°C or less, more preferably 25°C or less, from the viewpoint of the detergency. Especially, a polyoxyalkylene alkyl ether obtained by adding 6 to 10 moles of an alkylene oxide to an alcohol having 10 to 14 carbon atoms is preferable. Here, preferable as an alkylene oxide is ethylene oxide. In addition, the nonionic surfactant may be used in the form of an aqueous solution.

[0014]

The content of the nonionic surfactant is preferably from 25 to 99% by weight, more preferably from 30 to 95% by weight, of (b) component.

[0015]

The immobilization agent of the nonionic surfactant in (b) component means a component is capable of suppressing the flowability of the above nonionic surfactant at an ordinary temperature, for instance, at 25°C, enhancing the hardness of (b) component in a temperature range of less than a pour point of (b) component, and suppressing the viscosity of (b) component

to 10 Pa·s or less, in a temperature range higher than a pour point of (b) component by 10°C or more.

[0016]

The immobilization agent includes, for instance, anionic surfactants such as salts of fatty acids, salts of hydroxyfatty acids, and alkyl phosphates; polyoxyalkylene-type nonionic compounds and polyether-type nonionic compounds, such as polyethylene glycols, polypropylene glycols, and polyoxyethylene alkyl ethers; and the like. These compounds may be used alone or in combination of plural kinds.

[0017]

Since the immobilization agent is formulated, there are exhibited highly remarkable effects that the viscosity of (b) component is not increased in a temperature range higher than the pour point of (b) component, and that the hardness of (b) component can be markedly enhanced in the temperature range of less than the pour point of (b) component, whereby the penetrability of (b) component through (a) component in the former temperature range can be maintained, and the exudation of the nonionic surfactant in the latter temperature range can be effectively suppressed (hereinafter referred to as "immobilization ability").

[0018]

When the immobilization agent comprises an anionic surfactant, it is preferable that (b) component contains 5 to 25% by weight of water.

[0019]

The content of the immobilization agent in (b) component is from 1 to 100 parts by weight, preferably from 5 to 50 parts by weight, based on 100 parts by weight of the nonionic surfactant. Based on 100 parts by weight of the nonionic surfactant, the immobilization agent is preferably 1 part by weight or more, from the viewpoint of the immobilization ability of the nonionic surfactant, and the immobilization agent is preferably 100 parts by weight or less, from the viewpoint of the dissolubility of the detergent particles.

[0020]

It is preferable that (b) component substantially comprises no fatty acids. This feature enables to achieve an increase in the amount of (b) component supported to (a) component and an improvement in the dissolubility of the detergent particles. Here, the term "substantially comprises no fatty acids" refers to a case where a content of a fatty acid is 1% or less, and it is preferable that the fatty acid is undetectable, when (b) component is subjected to quantification of fatty acids by a method in accordance with Standard Fats and Oils Analysis Test

Method 2.4.1-71, Edited by Nippon Yukagaku Kyokai. The above effects are thought to be exhibited as follows. Concretely, when (b) component comprises a fatty acid, a salt of a fatty acid is formed by neutralization of the fatty acid with a component showing an alkaline property during mixing in step (I), whereby the salt of a fatty acid and the nonionic surfactant of (b) component are gelated. The formed gelated product inhibits the support of (b) component to (a) component, thereby lowering the supporting efficiency. In addition, a large aggregate is formed by the action of the gelated product as a binder, and (a) component is likely to undergo breakdown by applying a strong shearing force during mixing, and consequently making it disadvantageous in the dissolubility.

[0021]

The viscosity of (b) component is determined by measuring using a B-type viscometer (commercially available by TOKYO KEIKI, Model "DVM-B") under the conditions of rotor No. 3, at 12 rpm. The pour point of (b) component is determined by a method in accordance with JIS K 2269.

[0022]

It is preferable that (b) component further comprises an anionic surfactant having sulfate group or sulfo group.

The content of the anionic surfactant is preferably from 20 to 200 parts by weight, more preferably from 30 to 180 parts by weight, based on 100 parts by weight of the nonionic surfactant. Based on 100 parts by weight of the nonionic surfactant, the anionic surfactant is preferably 20 parts by weight or more, from the viewpoints of the suppression of the exudation of the nonionic surfactant and the improvement in the anti-caking ability, and the anionic surfactant is preferably 200 parts by weight or less, from the viewpoint of the dissolubility of the detergent particles. Since the anionic surfactant is formulated in (b) component, the exudation of the nonionic surfactant is further suppressed, and moreover, and the anti-caking ability of the detergent particles is improved, whereby detergent particles having desirable foaming ability and detergency performance can be obtained.

[0023]

Concrete examples of the anionic surfactant having sulfate group or sulfo group include linear alkylbenzenesulfonates, alkyl sulfates,  $\alpha$ -sulfonated fatty acid salts, polyoxyethylene alkyl ether sulfates, and the like.

[0024]

### 3. Powder Raw Materials Other Than (a) Component

In the present invention, powder raw materials other



than (a) component may be used. The term "powder raw materials other than (a) component" as referred to in the present specification means a detergency-enhancing agent or an oil-absorbing agent which is powdery at an ordinary temperature, for instance, 25°C. Concretely, the powder raw materials are base material agents exhibiting a metal ion capturing ability such as zeolite and citrates; base material agents exhibiting an alkalizing ability such as sodium carbonate and potassium carbonate; base material agents exhibiting both a metal ion capturing ability and an alkalizing ability such as crystalline silicates; amorphous silica and amorphous aluminosilicates exhibiting poor metal ion capturing ability but high oil-absorbing ability; powder surfactants, and the like. By using the above powder raw materials in combination with (a) component as desired, an increase in the amount of (b) component formulated and the reduction of the deposition of the mixture within the mixer can be achieved, and the improvement in detergency can also be achieved.

[0025]

The powder raw materials other than (a) component are mixed by formulating as desired in step (I). The formulation amount in this case is preferably from 1 to 30 parts by weight, more preferably from 3 to 20 parts by weight, especially preferably from 3 to 15 parts by weight,

based on 100 parts by weight of (a) component. The formulation amount, based on 100 parts by weight of (a) component, is preferably 1 part by weight or more, from the viewpoint of exhibiting the desired effects, and the formulation amount is preferably 30 parts by weight or less, from the viewpoint of the dissolubility.

[0026]

4. Fine Powder [(c) Component] of Step (II)

(c) Component is a powder which is formulated for the purpose of coating the surface of a mixture prepared by mixing (c) component together with the mixture obtainable in step (I), thereby improving the flowability of the resulting detergent particles, and those having high ion exchanging ability and high alkalizing ability are preferable from the viewpoint of the detergency.

Concretely, aluminosilicates are preferable. Aside from the aluminosilicates, inorganic fine powders of calcium silicates, silicon dioxide, bentonite, talc, clay, amorphous silica derivatives and silicate compounds such as crystalline silicate compounds are preferable. In addition, metal soaps of which primary particle has a size of 10  $\mu\text{m}$  or less can be similarly used.

[0027]

The fine powder of which primary particle has an average particle size of from 0.1 to 10  $\mu\text{m}$  is preferable,

from the viewpoints of an improvement in the coating ratio of the surface of the detergent particle and an improvement in the flowability of the detergent particles. The average particle size of the fine powder is measured by a method utilizing light scattering, for instance, by a particle analyzer (commercially available by Horiba, LTD.), or it is measured by a microscopic observation.

[0028]

The amount of the fine powder used, based on 100 parts by weight of the mixture obtainable in step (I), is preferably from 5 to 100 parts by weight or more, more preferably from 10 to 75 parts by weight or more, especially preferably from 10 to 50 parts by weight or more. The amount of the fine powder used is preferably 5 parts by weight or more, from the viewpoint of efficiency of surface coating, and the amount is preferably 100 parts by weight or less, from the viewpoint of the flowability.

[0029]

## 5. Process for Preparing Detergent Particles

The process in the present invention comprises Step (I) and Step (II).

### 5-1. Step (I)

[0029]

In step (I), (a) component is mixed with (b)

component, under conditions such that a maximum temperature of a mixture of (a) component and (b) component during the period of an initiation of mixing and a termination of mixing is a pour point of (b) component or higher, preferably higher than the pour point by 5°C or more, still more preferably higher than the pour point by 10°C or more. By mixing under the conditions described above, the breakdown of (a) component can be suppressed, thereby supporting (b) component to (a) component. Mixing is carried out with maintaining a temperature of a mixture of (a) component and (b) component during a period of an initiation of mixing and a termination of mixing at a pour point of (b) component or higher, preferably higher than the pour point by 5°C or more, still more preferably higher than the pour point by 10°C or more, from the viewpoint of more effectively exhibiting the above effects. In addition, the temperature of the mixture is preferably adjusted to 95°C or lower, more preferably 90°C or lower, from the viewpoint of the thermal stability of (b) component.

[0030]

Since (b) component has a state exhibiting flowability, not a hard paste or solid state, by adjusting the maximum temperature of the mixture to the pour point of (b) component or higher, (b) component can be easily

penetrated through (a) component by simply mixing together (a) component and (b) component under the above temperature conditions. Further, since (b) component is always in a state exhibiting the flowability described above throughout step (I) by mixing the components, with maintaining the temperature of the mixture at a pour point of (b) component or higher, (b) component can be very highly efficiently penetrated through (a) component. In addition, in a case where (b) component is in a state of hard paste or solid, a strong shearing force is acted to (a) component owing to the strong adhesiveness exhibited by (b) component, thereby giving rise to a possible breakdown of (a) component. The shearing force acting on (a) components themselves can be reduced by maintaining a temperature of the mixture at a pour point of (b) component or higher, whereby the breakdown of (a) component can be suppressed. Accordingly, from these aspects, it is preferable that the mixing operation is carried out in a state where (b) component exhibits flowability.

The temperature of the mixture is determined by an on-line measurement by setting a thermocouple at a position less likely to be influenced by a jacket or the like in the mixer.

[0031]

A preferable embodiment for satisfying the above temperature conditions is to initiate mixing after raising each of the temperatures of (a) component and (b) component to a pour point of (b) component or higher. In addition, in order to maintain the temperature of the mixture at a pour point or higher as mentioned above, it is preferable to previously adjust the jacket temperature to a pour point of (b) component or higher by allowing warm water or the like to flow through the jacket prior to mixing operation. The jacket temperature is preferably higher than the pour point by 5°C or more, especially preferably higher than the pour point by 10° or more. In addition, the jacket temperature is preferably 95°C or lower, more preferably 90°C or lower, from the viewpoint of the thermal stability of (b) component.

[0032]

As the method for adjusting a temperature of (a) component, when (a) component is obtained by spray-drying, it is preferable that the temperature of the particle immediately after spray-drying is usually a relatively high temperature, and that the particle is supplied in the mixer such that this temperature can be maintained. In addition, the temperature of the particle before or after supplying to the mixer can be previously heated by, for instance, a hot air.

[0033]

The mixing ratio of (a) component and (b) component is from 15 to 100 parts by weight or more, preferably from 25 to 80 parts by weight or more, more preferably from 30 to 70 parts by weight or more, based on 100 parts by weight of (a) component. (b) Component is preferably 15 parts by weight or more, from the viewpoint of exhibiting the detergency, and it is preferably 100 parts by weight or less, from the viewpoints of the dissolubility and the flowability, based on 100 parts by weight of the base particle.

[0034]

As the process for adding (b) component, a process comprising previously mixing each of ingredients constituting (b) component, i.e. a nonionic surfactant, an immobilization agent, and an anionic surfactant if used, and adding the mixture into the mixer is preferable.

[0035]

As to other mixing conditions, it is preferable that mixing is carried out with making the agitation force as small as possible within the mixable range of the (a) component and (b) component in order to suppress the breakdown of (a) component. When a mixer comprises agitation impellers, the agitation impellers have a Froude number of preferably 8 or less, more preferably from 4 or

less, still more preferably from 2 or less. The agitation impellers have a Froude number of preferably 0.5 or more, more preferably 0.8 or more, from the viewpoint of the efficiency of mixing. When a mixer comprises agitation impellers and disintegration impellers, it is preferable to mix only with the agitation impellers, without substantially rotating the disintegration impellers. The phrase "without substantially rotating the disintegration impellers" refers to completely not rotating the disintegration impellers at all, or rotating some of the disintegration impellers, for the purpose of preventing residence of various raw materials near the disintegration impellers, within a range such that the base particle does not substantially undergo breakdown, in consideration of shapes, sizes, and the like of the disintegration impellers. Concretely, in a case where the disintegration impellers are continuously rotated, the Froude number is 200 or less, preferably 100 or less, and in a case where the disintegration impellers are intermittently rotated, the Froude number is not particularly limited. The mixture can be obtained without substantially undergoing breakdown of the base particle by mixing under the conditions described above. In the present specification, a state where a base particle does not substantially undergo breakdown refers to a state such



that 70% or more of base particle in the mixture maintains its shape. Its method for confirmation includes, for instance, a method of subjecting to observation granules obtained after extracting a soluble fraction from a mixture obtained by using an organic solvent.

[0036]

The Froude number as defined in the present specification is calculated by the following equation.

$$\text{Froude Number} = V^2 / (R \times g),$$

wherein V: a peripheral speed [m/s] of a tip end portion of an agitating impeller or disintegration impeller;

R: a rotational radius [m] of an agitating impeller or disintegration impeller; and  
g: gravitational acceleration [m/s<sup>2</sup>].

[0037]

Further, in a case where (a) component is susceptible to undergo breakdown, (b) component may be supported to (a) component by arbitrarily adjusting a number of rotations of the agitation impellers (including stopping). Preferable mixing time (in the case of batch process) and average residence time (in the case of continuous process) are, for instance, preferably from 1 to 20 minutes, especially preferably from 2 to 10 minutes.

[0038]

In the case where mixing is carried out by a batch process, the mixer is not particularly limited, as long as a mixer which can satisfy the above condition is employed. Examples of the mixers include (1) a mixer in which blending of powders is carried out by having an agitating shaft in the inner portion of a mixing vessel and attaching agitating impellers on the agitating shaft, including Henschel Mixer (manufactured by Mitsui Miike Machinery Co., Ltd.); High-Speed Mixer (Fukae Powtec Corp.); Vertical Granulator (manufactured by Powrex Corp.); Lödige Mixer (manufactured by Matsuzaka Giken Co., Ltd.); PLOUGH SHARE Mixer (manufactured by PACIFIC MACHINERY & ENGINEERING Co., LTD.), and the like.

Examples of the mixers include (2) a mixer in which blending is carried out by rotating spiral ribbon impellers in a non-rotatable vessel which is cylindrical or semi-cylindrical, including Ribbon Mixer (manufactured by Nichiwa Kikai Kogyo K.K.); Batch Kneader (manufactured by Satake Kagaku Kikai Kogyo K.K.), and the like.

Examples of the mixers include (3) a mixer in which blending is carried out by revolving a screw along a conical vessel, with autorotation centering about a rotating shaft arranged parallel to the vessel wall, including Nauta Mixer (manufactured by Hosokawa Micron Corp.), and the like.

[0039]

Among the above mixers, a particular preference is given to a mixer in which blending of powders is carried out by having an agitating shaft arranged along the center line of a horizontal, cylindrical blending vessel and attaching agitating impellers arranged on the agitating shaft, including Lödige Mixer (manufactured by Matsuzaka Giken Co., Ltd.), PLOUGH SHARE Mixer (manufactured by PACIFIC MACHINERY & ENGINEERING Co., LTD.), and the like. Furthermore, it is preferable to use the mixers disclosed in Japanese Patent Laid-Open No. Hei 10-296065, because the temperature elevation of (a) component can be achieved by supplying hot air.

In a case where mixing is carried out in a continuous process, the mixer is not particularly limited, as long as a continuous mixer which can satisfy the present invention is employed. For instance, (a) component and (b) component may be mixed by using a continuous mixer among the above mixers.

[0040]

When the powder raw materials other than (a) component are formulated in step (I), it is preferable that the powder raw materials are supplied to the mixer before adding the surfactant composition. It is preferable that the mixing conditions when the powder raw

materials are formulated are the same conditions as those where (a) component and (b) component are mixed.

[0041]

In this step, the phrase "the state where (a) component does not substantially undergo breakdown" refers to a state in which 70% or more of (a) component in the mixture maintains its form. Its method for confirmation includes, for instance, a method of subjecting to SEM observation granules obtained after extracting a soluble fraction from a mixture obtained by using an organic solvent. When (a) component does not substantially undergo breakdown, there are advantages that the dissolubility and the flowability of the detergent particles are improved.

[0042]

#### 5-2. Step (II)

In this step, the mixture obtainable in step (I) is mixed with the fine powder [(c) Component]. In this step, the fine powder can coat the surface of the mixture, whereby detergent particles having excellent flowability can be obtained. In addition, when the mixture obtainable in step (I) does not show a powdery form (for instance, where (b) component forms a continuous phase, as in a case of a pasty or whipping form), step (II) includes a step for disintegrating the mixture using the fine powder as an aid.

[0043]

As the mixing conditions in this step, the condition that (a) component does not substantially undergo breakdown is preferable. Concretely, in the same manner as in step (I), the mixing is carried out under conditions that a maximum temperature of the mixed components of the mixture and the fine powder during an initiation of mixing and a termination of mixing is a pour point of (b) component or higher, preferably higher than the pour point by 5°C or more, still more preferably higher than the pour point by 10°C or more. From the viewpoint of further effectively exhibiting the above effects, the mixing is carried out with maintaining the temperature of the mixed components during an initiation of mixing and a termination of mixing at a pour point of (b) component or higher, preferably higher than the pour point by 5°C or more, still more preferably higher than the pour point by 10°C or more. In addition, from the viewpoint of the thermal stability of (b) component, the temperature of the mixed components is preferably 95°C or lower, more preferably 90°C or lower. A method for regulating the temperature of the mixed components during mixing includes a process comprising feeding warm water through the jacket of the mixer as in the same manner in step (I).

[0044]

In this step, the phrase "the state where (a) component does not substantially undergo breakdown" refers to a state in which 70% or more of (a) component in the detergent particle maintains its form. Its method for conformation includes a method of confirming the amount of the detergent particle constituted by one base particle by means of SEM observation.

[0045]

Other mixing conditions include those conditions using a mixer comprising both agitation impellers and disintegration impellers as preferred conditions, wherein the operating conditions (number of rotations, and the like) of the agitation impellers and the disintegration impellers may be properly set to an extent that (a) component does not substantially undergo breakdown. Preferable mixing conditions are the use of a mixer comprising both agitation impellers and disintegration impellers, and when the above mixer is used, the agitation impellers provided in the mixer have a Froude number of preferably 10 or less, more preferably 7 or less, from the viewpoint of the suppression of breakdown of the base particle. The Froude number is preferably 2 or more, more preferably 3 or more, from the viewpoints of the efficiencies in the mixing with the fine powder and in the dispersion of the fine powder. The disintegration

impellers have a Froude number of preferably 200 or more, more preferably 500 or more, from the viewpoints of the efficiencies in the mixing with the fine powder and in the dispersion of the fine powder. The Froude number is preferably 8000 or less, more preferably 5000 or less, from the viewpoint of the suppression of the breakdown of the base particle. If the Froude number is in this range, the detergent particles having excellent flowability can be obtained. The mixing time is preferably from 0.5 to 3 minutes or so.

[0046]

Preferable mixers include, among the mixers exemplified in step (I), those comprising both agitation impellers and disintegration impellers. When the mixers as described above are used, it is preferable from the viewpoint of simplification of equipments because the same device can be used in step (A) and step (B). The devices as described above include Lödige Mixer (manufactured by Matsuzaka Giken Co., Ltd.), PLOUGH SHARE Mixer (manufactured by PACIFIC MACHINERY & ENGINEERING Co., LTD.), and the like.

[0047]

## 6. Detergent Particles

The detergent particles in the present invention have a degree of particle growth of 1.5 or less, preferably 1.3

or less, more preferably 1.2 or less.

[0048]

In the above detergent particles, since the intraparticle aggregation is suppressed, the formation of the particle having a size outside the desired particle size range (aggregated particle) is suppressed (i.e. showing that variations in the average particle size and in the particle size distribution of the resulting detergent particles are made small against the variation in the amount of the surfactant formulated), so that detergent particles having excellent dissolubility are obtained in a high yield.

[0049]

#### 7. Preferable Properties Detergent Particles/Methods for Determining the Properties

The bulk density of the detergent particles is 500 g/L or more, preferably from 500 to 1,000 g/L, more preferably from 600 to 1,000 g/L, especially preferably from 650 to 850 g/L. The average particle size of the detergent particles is preferably from 150 to 500  $\mu\text{m}$ , more preferably from 180 to 350  $\mu\text{m}$ . The method for determining a bulk density and an average particle size is the same as that for (a) component.

[0050]

The flowability of the detergent particles is



evaluated as flow time of preferably 10 seconds or shorter, more preferably 8 seconds or shorter. The flow time is a time period required for dropping 100 mL of powder from a hopper used in the determination of the bulk density as defined in JIS K 3362.

[0051]

The anti-caking property of the detergent particles is evaluated as sieve permeability of preferably 90% or more, more preferably 95% or more. The testing method for caking property is as follows.

An open-top box having dimensions of 10.2 cm in length, 6.2 cm in width, and 4 cm in height is made out of a filter paper (No. 2, manufactured by ADVANTEC) by stapling the filter paper at four corners. An acrylic resin plate (15 g) and a lead plate (250 g) are placed on the box charged with a 50 g sample. The caking state after allowing the box to stand at a temperature of 35°C and a humidity of 40% for 2 weeks is evaluated by calculating the permeability as follows.

<Permeability> A sample obtained after the above test is gently placed on a sieve (sieve opening: 4760  $\mu\text{m}$ , as defined by JIS Z 8801), and the weight of the powder passing through the sieve is measured. The permeability (%) based on the sample obtained after the above test is calculated.

[0052]

As to the exudation property of the detergent particles, the evaluation by the following test methods is preferably 2 rank or better, more preferably 1 rank. When the exudation property is ranked as above, it is preferable because contrivances are not necessary for prevention of deposition of the nonionic surfactant-containing powder to equipments during transportation, or for prevention for exudation to vessels.

The testing method for exudation property: Exudation state of a surfactant is visually examined at bottom (side not contacting with powder) of the vessel made of the filter paper after the anti-caking test. The examination is made based on the area of wetted portion occupying the bottom in the following 1 to 5 ranks.

Rank 1: not wetted; Rank 2: about one-quarter of the bottom area being wetted; Rank 3: about one-half the bottom area being wetted; Rank 4: about three-quarter of the bottom area being wetted; Rank 5: the entire bottom area being wetted.

[0053]

The dissolution rate of the detergent particles is preferably 90% or more, more preferably 95% or more. The testing method for dissolution rate is as follows.

[0054]

A 1-L beaker (a cylindrical form having an inner diameter of 105 mm and a height of 150 mm, for instance, a 1-L glass beaker manufactured by Iwaki Glass Co., Ltd.) is charged with 1 L of hard water cooled to 5°C and having a water hardness corresponding to 71.2 mg CaCO<sub>3</sub>/L (a molar ratio of Ca/Mg: 7/3). With keeping the water temperature constant at 5°C, water is stirred with a stirring bar (35 mm in length and 8 mm in diameter, for instance, Model "TEFLON MARUGATA-HOSOGATA" manufactured by ADVANTEC) at a rotational speed (800 rpm), such that a depth of swirling to the water depth is about 1/3. The detergent particles are reduced and weighed so as to be 1.0000 ± 0.0010 g are supplied and dispersed in water with stirring, and stirring is continued. After 60 seconds from supplying the detergent particles, a liquid dispersion of the detergent particles in the beaker is filtered with a standard sieve (100 mm in diameter) and a sieve-opening of 74 µm as defined by JIS Z 8801 (corresponding to ASTM No. 200) of a known weight. Thereafter, water-containing detergent particles remaining on the sieve is collected in an open vessel of a known weight together with the sieve. Incidentally, the operation time from the start of filtration to collection of the sieve is set at 10 ± 2 sec. The insoluble remnants of the collected detergent particles are dried for one hour in an electric dryer

heated to 105°C. Thereafter, the dried insoluble remnants are cooled by keeping in a desiccator with a silica gel at 25°C for 30 minutes. After cooling the insoluble remnants, a total weight of the dried insoluble remnants of the detergent, the sieve and the collecting vessel is measured. Thereafter, the dissolution rate (%) of the detergent particles is calculated by the following equation. Incidentally, the weight is determined by using an accurate balance.

[0055]

$$\text{Dissolution Rate (\%)} = \{1 - (T/S)\} \times 100$$

wherein S is a weight (g) of the detergent particles supplied; and

T is a dry weight (g) of insoluble remnants of the detergent particles remaining on the sieve when an aqueous solution prepared under the above stirring conditions is filtered with the sieve, wherein drying conditions for the insoluble remnants are keeping at a temperature of 105°C for 1 hour, and then in a desiccator with a silica gel at 25°C for 30 minutes.

[0056]

The yield of the detergent particles is calculated from a weight percentage of a sample passing through a sieve having a sieve-opening of 1000  $\mu\text{m}$  when the average

particle size is determined. The yield is preferably 90% or more, more preferably 95% or more.

[0057]

[Examples]

The base particle used hereinbelow was prepared as described below.

Four-hundred and eighty kilograms of water was supplied to a 1-m<sup>3</sup> mixing vessel comprising agitation impellers. After the water temperature reached 55°C, 120 kg of sodium sulfate and 150 kg of sodium carbonate were added thereto. The mixture was stirred for 15 minutes, and thereafter 120 kg of a 40% by weight aqueous solution of sodium polyacrylate was added thereto. The resulting mixture was stirred for additional 15 minutes, and thereafter 252 kg of zeolite was added thereto. The resulting mixture was stirred for 30 minutes, to give a homogenous slurry. The final temperature of this slurry was 58°C. This slurry was subjected to spray-drying, and the resulting spray-dried particles were used as a base particle. The base particle had an average particle size of 245 µm, a bulk density of 610 g/L, a supporting ability of 50 mL/100 g, a particle strength of 350 kg/cm<sup>2</sup>, and a composition (weight ratio) of zeolite/sodium polyacrylate/sodium carbonate/sodium sulfate/water of 42/8/25/20/5.

[0058]

Example 1

Detergent particles were obtained according to the following process.

One-hundred parts by weight (20 kg) of a base particle at 80°C as listed in Table 1 was supplied into Lödige Mixer (commercially available from Matsuzaka Giken Co., Ltd.; capacity: 130 L; equipped with a jacket), and the rotation of a main shaft (rotational speed: 60 rpm) was started. Incidentally, hot water at 80°C was allowed to flow into the jacket at 10 L/minute, without rotating a chopper. Forty-five parts by weight (9 kg) of a surfactant composition at 80°C was supplied into the above mixer over a period of 2 minutes, and thereafter the components were mixed for 5 minutes. The temperature of the mixture immediately after introducing the surfactant was 73°C, and the temperature of the mixture after stirring for 5 minutes was 74°C.

Subsequently, 15 parts by weight (3 kg) of fine powder was supplied into this mixer, with continuing to feed hot water to the jacket. The main shaft (rotational speed: 120 rpm) and the chopper (rotational speed: 3600 rpm) were rotated for 1 minute, and thereafter 30 kg of detergent particles were discharged. The properties of the obtained detergent particles are shown in Table 1.

[0059]

[Table 1]

Table 1

	Examples		Comp.
	1	2	Ex. 1
<u>Composition (Parts by Weight)</u>			
Surfactant Composition			
Polyoxyethylene Alkyl Ether	40	20	40
Polyethylene Glycol	5	2	5
Sodium Dodecylbenzenesulfonate	-	20	-
Water	-	3	-
Base Particle (Spray-Dried Particle)	100	100	100
Fine Powder (Zeolite 4A, average particle size of 3.5 $\mu\text{m}$ )	15	15	15
<u>Properties</u>			
Pour Point ( $^{\circ}\text{C}$ ) of Surfactant Composition	52.5	47.5	52.5
Maximum Temperature ( $^{\circ}\text{C}$ ) of Step (I) Mixture	74	72	45
Minimum Temperature ( $^{\circ}\text{C}$ ) of Step (I) Mixture	73	68	40
Maximum Temperature ( $^{\circ}\text{C}$ ) of Step (II) Mixture	72	70	44
Minimum Temperature ( $^{\circ}\text{C}$ ) of Step (II) Mixture	68	65	41
Average Particle Size ( $\mu\text{m}$ )	252	277	409
Degree of Particle Growth	1.03	1.13	1.67
Bulk Density (g/L)	780	790	820
Exudation Property (2-Week Storage)	2-3	2	2
Anti-Caking Property (2-Week Storage)	2-3	2	2
Dissolution Ratio (%)	97	95	85
Yield (%)	99	97	88

[0060]

Here, as the polyoxyethylene alkyl ether, there was used one commercially available from Kao Corporation under the trade name of "EMULGEN 108 KM" (average moles of ethylene oxides: 8.5; number of carbon atoms in alkyl moiety: 12 to 14; melting point: 18°C). As the polyethylene glycol, there was used one commercially available from Kao Corporation under the trade name of "K-PEG 6000" (average molecular weight: 8500; melting point: 60°C). As sodium dodecylbenzenesulfonate, there was used one commercially available from Kao Corporation under the trade name of "NEOPELEX FS."

[0061]

Example 2

Detergent particles were obtained in the same manner as in Example 1 with the composition listed in Table 1. The temperature of the mixture immediately after supplying the surfactant was 72°C, and the temperature of the mixture after stirring for 5 minutes was 68°C. The properties of the obtained detergent particles are shown in Table 1. The detergent particles of Example 2 were more excellent in the anti-caking property and the exudation property than the detergent particles of Example 1.

[0062]

From the finding that both of the final detergent



particles obtained in Examples 1 and 2 were low in the degree of particle growth, they were uni-core detergent particles. In addition, as a result of observing the particles after extracting and removing soluble matter from the mixture obtained after the completion of step (I) and the final detergent particles using an organic solvent, the base particles in the mixture and the base particle in the detergent particles did not substantially undergo breakdown in each of Examples 1 and 2.

[0063]

#### Comparative Example 1

Detergent particles were obtained in the same manner as in Example 1, except for the temperature of the base particle and the temperature of hot water in the jacket. Specifically, the temperature of the base particles when supplied was 25°C, and the temperature of water flowing into the jacket was 25°C. The temperature of the mixture immediately after supplying the surfactant was 45°C, and the temperature of the mixture after stirring for 5 minutes was 40°C.

[0064]

Subsequently, 15 parts by weight (3 kg) of fine powder was supplied into this Lödige Mixer. The main shaft (rotational speed: 120 rpm) and the chopper (rotational speed: 3600 rpm) were rotated for 1 minute,

and thereafter 27 kg of detergent particles were discharged. The properties of the obtained detergent particles are shown in Table 1. Since the degree of granule growth was large, the obtained detergent particles were not uni-core detergent particles. In addition, their dissolubility was poor. As a result of observing the particles after extracting and removing soluble matter from the mixture obtained after the completion of step (I) and the final detergent particles using an organic solvent, the average particle sizes of the base particle were reduced to about 50% for the mixture, and reduced to about 40% for the final detergent particles.

[0065]

[Effects of the Invention]

Further, according to the process of the present invention, there can be obtained detergent particles capable of having a large formulated amount of the surfactant, obtainable in a simplified preparation steps, excellent in the dissolubility, and excellent in the exudation suppression and the anti-caking property of the nonionic surfactant.